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CITATION:

Mori, Shigeo. Phase separation and ferromagnetic microdomains in doped manganites. 物性研究 2002, 79(2): 187-188

ISSUE DATE:

2002-11-20

URL:

http://hdl.handle.net/2433/97369

Phase separation and ferromagnetic microdomains in doped manganites

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Manganites with the perovskite structure exhibit a variety of unusual physical properties such as a colossal magnetroresistance (CMR) and a metal to insulator (MI) transition. Recent theoretical and experimental studies pointed out that the spatial inhomogeneous state of magnetic, electric and/or lattice systems, which is characterized as phase separation, is important to understand unusual physical properties in the doped-manganites.[1] Recently it was found that, by substituting Cr ions for the Mn ones in doped-manganites, the long-ranged charge and orbital ordered (CO) state becomes short-ranged one and new ferromagnetic (FM) metallic state is induced.[2] In addition, Kimura et al. found that the impurity-doped manganites ($Nd_{0.5}Ca_{0.5}Mn_{1-y}Cr_yO_3$) show characteristic physical properties similar with *relaxor* ferroelectrics such as $Pb(Nb_{1/3}Mg_{2/3})O_3$. [3] These anomalous properties found in the impurity-doped manganites have close relation to spatially inhomogeneous state of the FM state and the CO one. Here we reported structural change related to the anomalous properties found in the impurity-doped manganites.[4] In particular, we investigated the microstructure related to both the CO state and the FM one with a spatial resolution of ~nm by means of both the transmission electron microscopy and low-temperature Lorentz microscopy.

The magnetic and electric properties in the impurity-doped $Nd_{0.5}Ca_{0.5}MnO_3$ were shown in Fig.1. As is understood in Fig.1, it is found that the magnetic moment is induced by small amount of the Cr doping. That is, the Cr-doping in $Nd_{0.5}Ca_{0.5}MnO_3$ induced new FM state below the low temperature of 150K. In order to clarify the effect of the impurity substitution for the Mn sites in $Nd_{0.5}Ca_{0.5}MnO_3$, we examined carefully change in the physical properties by substituting the Sc ion for the Mn one. Temperature dependence of the magnetization (M) in $Nd_{0.5}Ca_{0.5}Mn_{0.97}Sc_{0.03}O_3$ was also shown in Fig.1. A definite cusp structure, which corresponds to the antiferromagnetic CO transition, is found around 60K.

We investigated the effect of the impurity-doping on the microstructure related to the CO state by obtaining both electron diffraction (ED) patterns and dark-field (DF) images. It was found from the ED experiments that the Cr-doped $Nd_{0.5}Ca_{0.5}MnO_3$ has a superlattice structure with the incommensurate wave vector of $q=(1/2-\delta)a_o^*$. Figures 2(a)-2(d) show DF images taken by using the superlattice spots characterizing the CO structure. As shown in Figs.2(a)-2(d) obtained at 90K, it is revealed that the CO state exists as microdomains in $Nd_{0.5}Ca_{0.5}Mn_{1-y}Cr_yO_3$ for y=0.01, 0.02, and 0.03. The size of the CO state is estimated to be about 10-30nm, where the size of the CO microdomains is strongly affected by the amount of the Cr substitution. As is evident from Fig.2, the low-temperature phase in $Nd_{0.5}Ca_{0.5}Mn_{1-y}Cr_yO_3$ is characterized as the mixed state of the CO state and the FM one. On the other hand, we found the presence of the incommensurate (IC)-tocommensurate (C) transition around 60K in $Nd_{0.5}Ca_{0.5}Mn_{0.97}Sc_{0.03}O_3$. Thus, we investigated thoroughly structural change related to the IC-to-C phase transition. Figure 3(a) shows the microstructure obtained around 186K. The CO state appears as the microdomains with the size of 10-20nm. On cooling down the sample, the CO microdomains grows up at the expense of the charge and orbital disordered regions, as shown Figs.3(b) and 3(c). In the cooling process down to 13K, we found the change from the CO microdomains to the large CO domains around 60K, as shown in Figure 3(d). That is, the IC-to-C structural phase transition accompanies the formation of the long-ranged CO structure.

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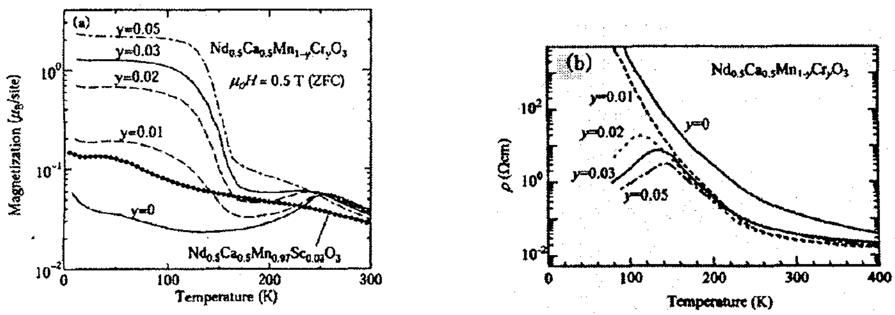

Figure1. Cr concentration dependence of (a) magnetization (M) and (b) resistivity (ρ) for $Nd_{0.5}Ca_{0.5}MnO_3$. Magnetization curve (M) for $Nd_{0.5}Ca_{0.5}Mn_{0.97}Sc_{0.03}O_3$ is also shown in (a).

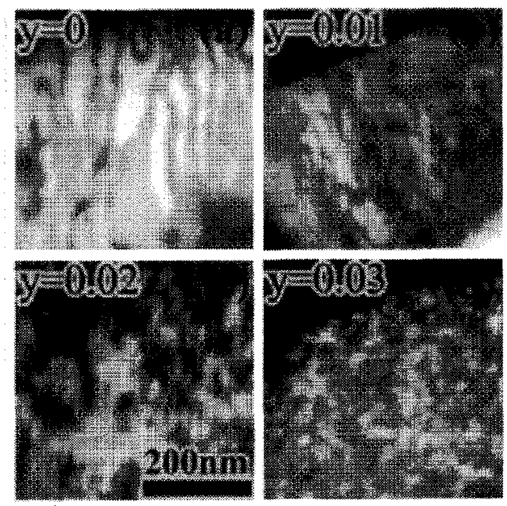

Figure2. Microstructure related to the CO state in $Nd_{0.5}Ca_{0.5}Mn_{1-y}Cr_yO_3$ with y=0.0(a), 0.01(b), 0.02(c), and 0.03(d), respectively.

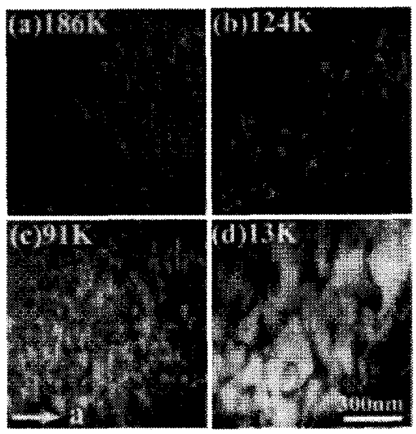

Figure3. Change in the microstructure related to the CO state in $Nd_{0.5}Ca_{0.5}Mn_{0.97}Sc_{0.03}O_3$, obtained in the cooling process from RT.